

(12) **UK Patent Application** (19) **GB** (11) **2 191 715** (13) **A**
 (43) Application published 23 Dec 1987

(21) Application No 8713004

(22) Date of filing 3 Jun 1987

(30) Priority data

(31) 875127 (32) 17 Jun 1986 (33) US

(71) Applicant

Midrex International B.V. Rotterdam Zurich Branch,

(Incorporated in Netherlands),

Wilfriedstrasse 12, 8032 Zurich, Switzerland

(72) Inventor

Glenn E. Hoffman

(74) Agent and/or Address for Service

Baron & Warren, 18 South End, Kensington,
 London W8 5BU

(51) INT CL⁴
 C10K 1/00

(52) Domestic classification (Edition I)

B1T FAA
 C5E 191 PG
 U1S 1360 B1T C5E

(56) Documents cited

GB A 2137115 US 4035170
 GB A 2024041 US 3928532
 GB 1586266 US 3632305

(58) Field of search

B1T
 B1W
 C5E
 Selected US specifications from IPC sub-classes B01D
 C10K

(54) **Method and apparatus for dedusting and desulfurizing gas**

(57) Hot fuel gas is fed upwards through a descending bed of refractory particles in a first vessel 22, which removes dust from the gas, and is then fed through a similar vessel 32 containing a descending bed of reactive material, including limestone, which removes sulphur.

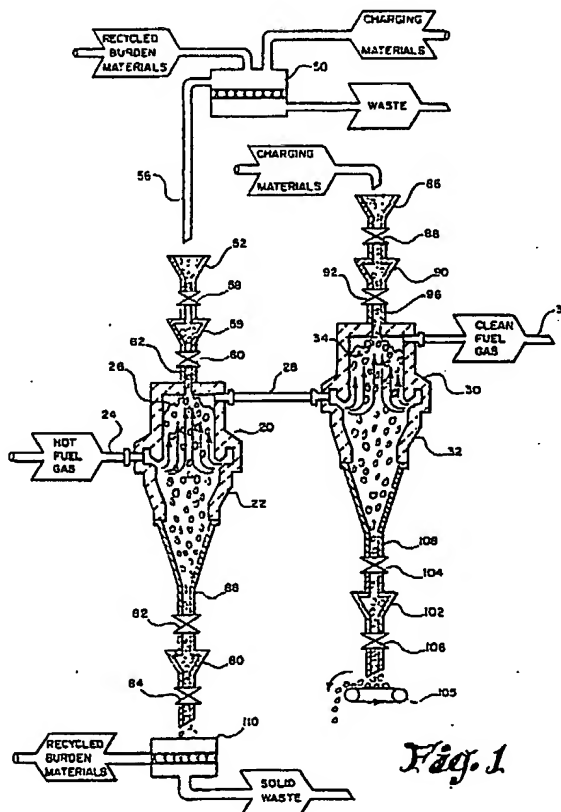
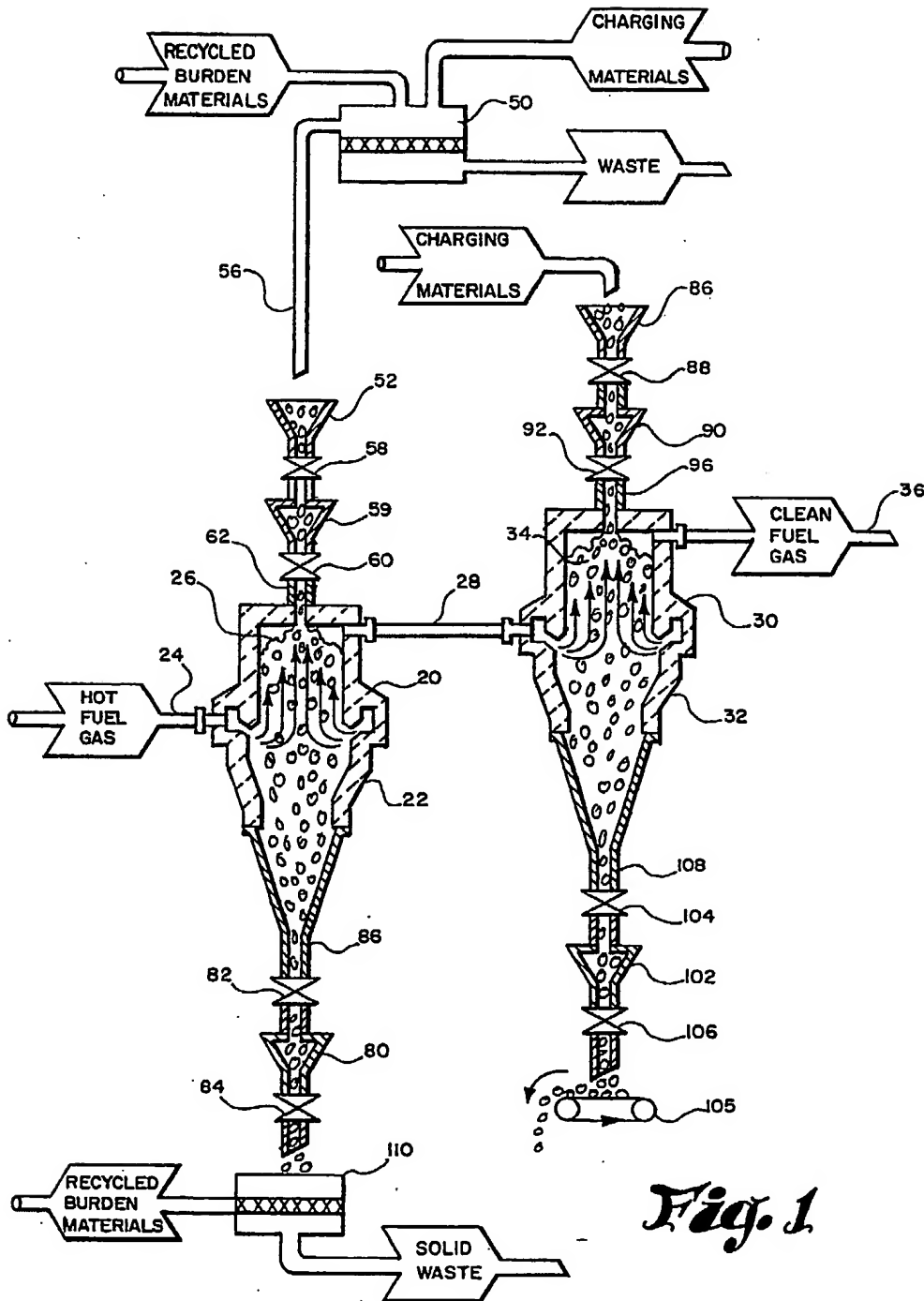


Fig. 1

GB 2 191 715 A



2191715

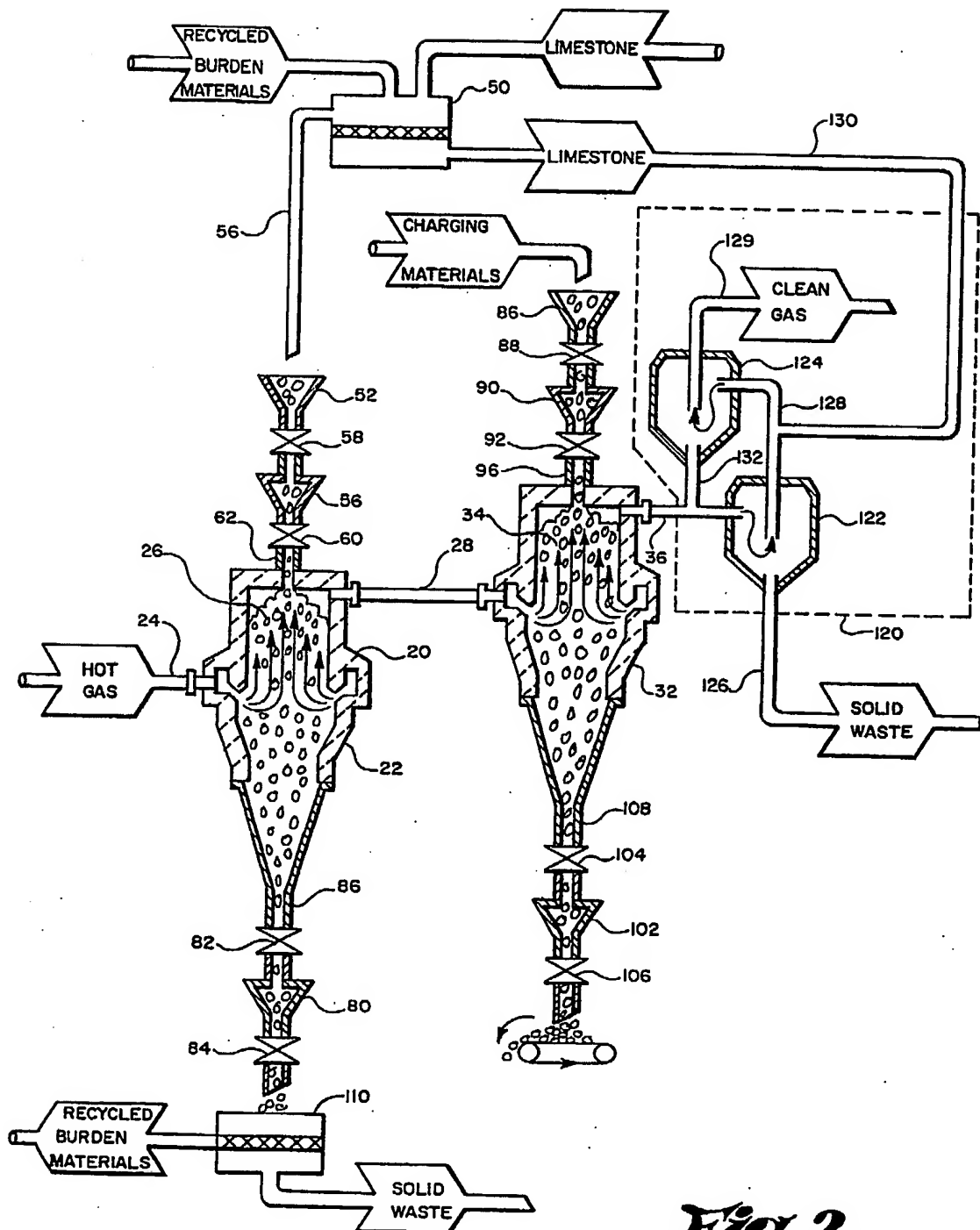


Fig. 2

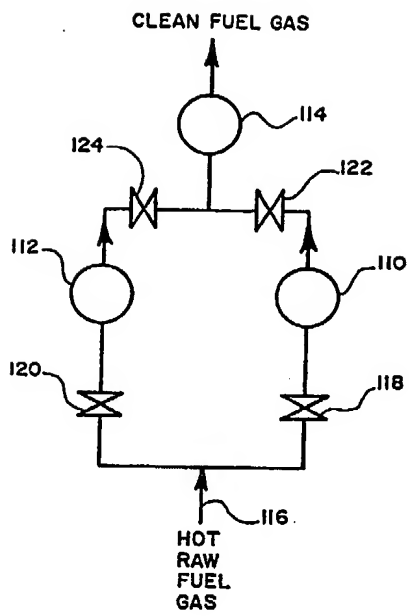


Fig. 3

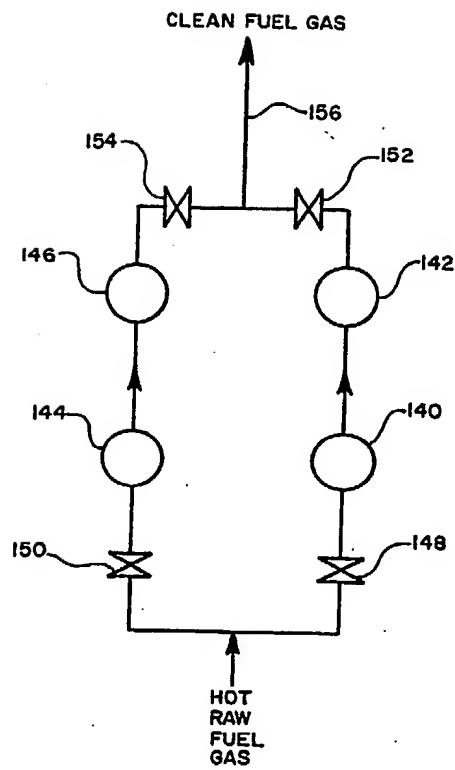


Fig. 4

SPECIFICATION

Method and apparatus for dedusting and desulfurizing gases5 *Background of the invention*

The present invention relates to a method and apparatus for removing solids and sulfur from hot gases.

- 10 Methods and systems for dedusting and desulfurizing fuel gases are well known in the art. It is known, for example, to utilize cyclone cleaners to remove dust and solids from synthetic gas produced by gasification of coal. Utilizing cyclone cleaners for
- 15 dust removal requires that the temperature of the fuel gas not exceed 1150°C, which is the upper temperature limit of available cyclones. Typically, the temperature of fuel gas produced by gasifying fossil fuels, such as coal, exceeds the operating
- 20 temperature range of typical hot cyclones. Additionally, it is well known that fuel gases containing sulfur and solids can be desulfurized and the solids removed by introducing the fuel gas into a shaft reactor in counterflow relationship to a burden
- 25 which can include such materials as limestone and iron oxide.

- Gasifiers utilizing coal generate fuel gases usually having a high concentration of solids and dust. Introducing gas streams having a high concentration
- 30 of dust and solids into shaft furnace reactors such as the Midrex Shaft Reactor for Producing Metallized Iron, may result in a variety of undesirable conditions including a high probability of accumulation of dust, which "plugs" the burden
- 35 including a high probability of accumulation of dust, which "plugs" the burden preventing the normal counterflow between the burden and the fuel gas, and impedes normal burden descent through the furnace. As a result, techniques and apparatus for
- 40 removing dust interposed between the gasifier and the shaft reactor are desirable.

- Interposing hot cyclones between the gasifier and the shaft reactor is one way of removing such solids and dust. Such a technique is illustrated in U.S.
- 45 Patent Number 4,280,412. The use of hot cyclones may require precooling of the hot fuel gas prior to introduction into the cyclone. In order to achieve high efficiency in a cyclone, particulate velocities are extremely high, with the result that the particulates
- 50 cause high erosion of the cyclone wall, and often erosion of the cyclone throat. At extremely high temperatures (above 1150°C), high temperature refractories are required, which do not resist high particulate velocities well. Thus, high erosion of the
- 55 cyclone wall and throat continues to be a problem.

- As taught by this invention, solids tend to adhere to the surface of the burden materials or solids are entrapped in the bed interstices to provide removal of a portion of the solids. Shaft reactors may be used
- 60 for gross solids removal using a burden consisting primarily of refractory materials which are recirculated while hot. This reduces energy losses. The partially cleaned fuel gas is introduced into a second shaft reactor and reacts with the burden
- 65 therein to remove sulfur and additional solids. If

desired, the fuel gas leaving the second shaft reactor and fine limestone may be introduced into a hot cyclone to provide additional desulfurization and cleaning.

- 70 Synthetic fuel gases have also been precooled using various techniques. Typically, the hot fuel gas has been mixed with a cooler tempering gas to lower its temperature sufficiently to permit the solids and sulfur to be conveniently removed. Generally, these
- 75 techniques adversely impacted the overall efficiency of the system in that the temperature reducing technique invariably resulted in an irrecoverable energy loss.

- Shaft dust removal systems have a longer life expectancy than hot cyclones, as the gas and particulate velocities are substantially lower, which allows higher temperature gas introduction to the shaft without concomitant erosion of refractories and walls of the shaft.

85 *Summary of the invention*

- This invention provides apparatus and a method for removing solids and sulfur from a hot fuel gas. Gross solid removal (removal of the larger solids and
- 90 a portion of the smaller solids) is provided by introducing hot fuel gas into a first shaft reactor charged with refractory pellets, large limestone or dolomitic limestone particles, or a combination thereof, to form a burden. As the hot fuel gas flows
- 95 through the burden, solids in the form of particulates, dust, and alkaline substances accumulate on the surface of the materials comprising the burden and in the burden interstices. A portion of the burden is periodically discharged
- 100 and replenished, removing the solids and alkaline substances which either adhere to the burden materials or are trapped in the interstices between the solids forming the burden.

- Hot fuel gas from the first shaft reactor is
- 105 introduced into a second shaft reactor which is charged with a material that includes limestone. As the hot fuel gas passes upwardly through the burden in the second shaft reactor, portions of the limestone are calcined. Sulfur components of the fuel gas
- 110 reacts with the calcined limestone to form CaS, thereby removing sulfur from the fuel gas stream. Additionally, solids carried over from the first shaft reactor tend to adhere to the surface of the calcined limestone and other materials comprising the
- 115 burden. Portions of the burden, including the limestone, calcined limestone, CaS and other burden materials are discharged from the second shaft reactor as the burden is replenished in accordance
- 120 with a predetermined schedule thus removing the sulfur and the solids from the second shaft reactor and the first gas stream. (Other burden materials which include sulfur removing agents such as dolomite may also be used.)

- As the limestone burden in the second shaft
- 125 reactor moves, dust may be liberated and carried over into the output of this reactor. As an option, a two-stage hot cyclone is provided to remove additional dust and other solids. Additional desulfurization may also be accomplished by
- 130 introducing fine limestone or other sulfur reducing

agents into these cyclones.

More specifically, the preferred embodiment of the invention comprises apparatus and a method for removing solids and sulfur from a hot fuel gas stream using a two-stage process in which the hot fuel gas is introduced into a shaft reactor in counterflow relationship to a burden including refractory pellets, large limestone particles or a combination thereof to remove solids and alkalis. After passing through the first shaft reactor, the hot fuel gas is introduced into a second shaft reactor in counterflow relationship to a second burden which includes calcined limestone. Sulfur containing components of the fuel gas interact with the calcined limestone to form solids, thus removing the sulfur from the fuel gas stream. Additional desulfurization may be provided by introducing the hot fuel gas stream from the second furnace and fine limestone into a hot cyclone.

Objects of the invention

It is an object of this invention to provide an economical and energy efficient method for removing solids and sulfur from a hot fuel gas. It is another object of this invention to provide a method and apparatus for removing sulfur and solids from a fuel gas utilizing shaft reactors. It is also an object of this invention to provide an efficient and economical method and apparatus for removing solids and sulfur from a hot fuel gas in which larger solids are removed by introducing the hot fuel gas into a first shaft reactor charged with refractory materials and the sulfur is removed by introducing the hot fuel gas into a second shaft reactor which is charged with a burden that includes calcined limestone.

Brief description of the accompanying drawings

Figure 1 is a schematic diagram of apparatus for cleaning and desulfurizing a hot fuel gas in which a hot fuel gas to be cleaned and desulfurized flows in a stream through first and second shaft reactors, the first primarily removing large solids, the second primarily removing sulfur.

Figure 2 is a schematic diagram of a second embodiment of the invention illustrated in Figure 1 which includes additional sulfur removing apparatus.

Figure 3 is a flow diagram illustrating a third embodiment of the invention using three shaft reactors permitting the burden in the first shaft reactor to be maintained static during use.

Figure 4 is a flow diagram illustrating a fourth embodiment of the invention in which the burdens in all of the reactors are maintained static during use.

Detailed description of the invention

Referring now to the embodiment of the invention illustrated in Figure 1, a hot fuel gas stream is introduced through a bustle and tuyere system 20 into a first shaft reactor 22 from a suitable input pipe 24. The hot fuel gas flows upwardly through a packed bed burden 26 within the reactor, which includes refractory pellets, limestone, or a mixture of refractory pellets and limestone. After passing

through the burden 26, the fuel gas flows through an outlet pipe 28 and is introduced through a bustle and tuyere system 30 into a second reactor furnace 32, containing burden 34. Solids, as well as interaction with the fuel gas, contaminate the materials of burdens 26 and 34 as the fuel gas flows therethrough. This contamination necessitates the periodic replenishment of the materials comprising the burdens 26 and 34.

More specifically, in the first reactor furnace 22, solids, such as dust and alkalis, adhere to the surfaces of the refractory pellets or large limestone particles for removal as the burden 26 is replenished. Partially cleaned fuel gas from the first reactor 22 flows through pipe 28 as discussed above and into the bustle and tuyere system 30 of the second shaft reactor 32. A second burden 34 in the shaft reactor 32 includes calcined limestone, which reacts with the sulfur components of the fuel gas to form solid CaS. Additional solids, not removed by or carried over from the first reactor 22, will also tend to adhere to the calcined limestone comprising the burden 34. These solids and the CaS are removed as the burden 34 is replenished. Hot clean fuel gas exits from the second shaft reactor 32 through a suitable pipe 36 for use, as desired. In order to assure that the hot fuel gas flows through the first and second reactors 22 and 32 at the desired rate and quantities, it is necessary that the system be pressurized.

In charging the first reactor 22, hot recycled refractory pellets, and alternatively limestone, are preferably fed to a screening station 50. In order to flow conveniently through the first shaft reactor 22, the burden materials must be larger than a preselected size, because high levels of fine particulate matter in a mixed size burden tends to clog shaft reactors and impede burden movement by promoting "bridging". Properly sized refractory pellets, and limestone, if desired, from the screening station 50 are fed into a feed hopper 52 through a suitable pipe 56.

A charging chamber 59 is positioned between first and second charging gates 58 and 60. Feed hopper 52, charging gate 58, charging chamber 59, and charging gate 60 are supported by a feed pipe 62 and attached thereby to the upper end of the first shaft reactor 22. A discharge chamber 80 is positioned between first and second discharged gates 82 and 84, with discharge gate 82 in turn being attached to the discharge pipe 86 of the first particle reactor 22.

A second charging chamber 90 is positioned between third and fourth charging gates 88 and 92. Support for the feed hopper 86, charging gates 88 and 92, and charging chamber 90 is provided by a feed pipe 96 attached to the upper end of the shaft reactor 32. A discharge chamber 102 is positioned between third and fourth discharge gates 104 and 106, and attached to the lower end of the shaft reactor 32 by a discharge pipe 108.

The initial burdens 26 and 34 in the first and second shaft reactors 22 and 32 can be established by closing the first, second, third and fourth discharge gates 84, 86, 106 and 108, opening the first, second, third and fourth charging gates 58, 60, 88, and 92, and pouring suitable quantities and types of

charging materials into feed hoppers 52 and 86. During operation, the charge and discharge gates must be operated in a sequence which permits the burdens 26 and 34 to be replenished, while the first and second shaft reactors remain pressurized.

For example, in replenishing the burden 26 in the first reactor 22, charge gate 60 is closed, sealing the upper end of the vertical reactor 22 to maintain pressure therein. Hot refractory pellets and other suitable burden materials from the screening station 50 are fed into the feed hopper 52 by pipe 56. The upper charging gate 58 is opened, permitting the burden materials to flow by gravity into charging chamber 59. After the charging chamber 59 is filled to a suitable level, the upper charging gate 58 is closed and the lower charging gate is opened, permitting burden materials to flow from the charging chamber 59 into the first reactor 22 to replenish the burden 26.

Portions of the burden 26 are discharged through the discharge pipe 86, upper discharge gate 82, discharge chamber 80 and lower discharge gate 84, in a similar fashion. That is, to discharge a portion of the burden 26, the lower discharge gate 84 is closed and the upper discharge gate 82 is opened, permitting burden materials to flow through the discharge pipe 86 and fill the discharge chamber 80. After discharge chamber 80 is filled, upper discharge gate 82 is closed and the lower discharge gate 84 is opened, permitting portions of the burden 26 to be discharged from the discharge chamber 80 into the recycling screening station 110, wherein hot refractory pellets are separated from the other burden materials. The hot refractory pellets are recycled to the first screening station 50 to be used as burden materials to replenish the burden 26. The remainder of the discharged burden materials are removed from shaft furnace 22 as solid waste.

Due to the requirement for continuously maintaining the shaft reactors 22 and 32, pressurized burden 26 cannot continuously flow downward through the first shaft reactor 22. This is because the upper discharge gate 82 must at least be closed during the period when the discharge chamber 80 is being emptied. Closing this discharge gate prohibits discharge of the burden materials through the discharge pipe 86. However, the periods during which this gate is closed may be selected to be sufficiently short that the burden 26 essentially moves continuously downward through the first reactor 22. Alternatively, the burden 26 can remain static as will be subsequently described.

Burden 34 in the second reactor 32 is replenished by utilizing a charging sequence similar to that described with reference to the first reactor 22. That is, the upper charging gate 88, charging chamber 90, and lower charging gate 92 are operating in a sequence similar to the charging process previously described for the first reactor 22. Similarly, portions of the burden 34 are removed from the second reactor 32 utilizing the discharge chamber 102 and upper and lower discharge gates 104 and 106 in a fashion very similar to that previously described with reference to the first vertical reactor 22 and discharge gates 82, 84 in conjunction with discharge chamber

80. Burden materials discharged from the second shaft reactor 32 in replenishing burden 34 are removed and discharged as solid waste by a suitable conveyor 105.

As previously described, the first vertical reactor 22 is charged using refractory material recycled from the screening station 110 through screening station 50 and feed hopper 52. Similarly, the second vertical reactor 32 is charged with either limestone or a mixture of iron oxide and limestone by supplying these burden materials to the feed hopper 86 and utilizing the feed gates 88 and 92 and feed chamber 90 to maintain the second vertical reactor 32 in a pressurized state while the burden 34 is being replenished.

As described above, the first vertical reactor 22 may be charged with refractory pellets, large limestone particles or a mixture thereof, and the second vertical reactor 32 charged with burden materials which include limestone to remove solids and sulfur from the hot fuel gas. As an alternative, the second vertical reactor 32 may be charged with limestone, dolomitic limestone or a mixture thereof, or a mixture of iron oxide and limestone and/or dolomite. If a charge consisting of a mixture of iron oxide and limestone is used in the second vertical reactor 32, the fuel gas introduced into the second vertical reactor 32 by way of the bustle and tuyere system 30 will react with the iron oxide to produce metallized iron. Under these conditions, the burden material discharged from the second vertical reactor 32 will consist of a mixture of calcium sulfide, metallized iron and other solid materials. These materials can be readily separated from each other by utilizing magnetic techniques to recover the metallized iron.

The sulfur-removing agent in the second reactor has an average particle diameter of at least 0.5 centimeters. If the particle size is too small, the bed will fluidize. The preferred average particle diameter is less than 3.0 centimeters. A limestone (or dolomitic limestone) average particle diameter of at least 0.5 centimeters is desirable to effect high utilization of the lime to form calcium sulfide, as well as requiring short residence times of the burden in the shaft reactor, which is amenable to high levels of dust removal from the gas stream. An average particle diameter greater than 3 centimeters renders the process uneconomical, as short burden residence times required for good dust removal result in low levels of lime utilization.

The alternative embodiment of Figure 2 is identical to the embodiment described in Figure 1, with the exception that additional apparatus for removal of solids and desulfurization is provided. This additional apparatus, enclosed within dotted line 120, includes first and second cyclones 122 and 124. Hot fuel gas flows from the top of the second vertical reactor 32 to the first cyclone 122 through discharge pipe 36. Solids removed by either cyclone 122 or 124 are ultimately discharged by a waste discharge pipe 126 and discarded as solid waste.

The gaseous output of the first cyclone 122 flows through a pipe 128 to the input of the second cyclone 124. Clean and desulfurized fuel gas from the second

cyclone 124 flows through an exit pipe for further use, as desired.

Fine particulate solids, including limestone, from the screening stage 50 are coupled by a suitable pipe 130 to the input of the second cyclone 124. Portions of this limestone combine with the remaining sulfur contained in the fuel gas to form solid CaS. The CaS, excess limestone, and other solids are discharged through discharge pipe 132 and combined with gaseous output of the second vertical reactor 32 to form the input materials to the first cyclone 22. These solids are ultimately removed from the fuel gas stream by the first cyclone 122, as previously described.

An embodiment of the invention which permits the burden materials in the first shaft reactor to remain static during the time that the fuel gas is flowing therethrough is illustrated by Figure 3. This embodiment utilizes three shaft reactors, 110, 112, and 114. Raw fuel gas is introduced through a pipe and alternatively directed to either shaft reactor 110 or 112 through valves 118 or 120. Fuel gas leaving the shaft reactors 110 and 112 is coupled through third or fourth valves 122 or 124 to the input of the third shaft furnace 114.

In operation, the third shaft furnace 114 is operated in a manner identical to the second shaft reactor 32, previously described with reference to Figure 1. The first and second shaft reactors of Figure 3, reactors 110 and 112, are charged with materials identical to shaft furnace 22, with the fuel gas being directed through these shaft reactors through the valves 118 and 120 in such manner that no fuel gas flows through the shaft reactor while its burden is being replenished. This reduces dust carryover during periods when the burdens are being replenished.

The embodiment of the invention illustrated in Figure 4 utilizes four shaft reactors 140, 142, 144, and 146. Raw fuel gas is introduced into shaft reactors 140 and 144 through first and second valves 148 and 150. Fuel gas leaves the shaft reactor 142 and 146 through valves 152 and 154 and pipe 156. In operation, shaft reactors 144 and 146 are operated in a manner identical to furnaces 22 and 32 to clean and desulfurize fuel gas, while shaft reactors 140 and 142 are being recharged to replenish their burdens. Similarly, shaft reactors 140 and 142 are operated to clean and desulfurize fuel gas while shaft reactors 144 and 146 are being recharged to replenish their burdens.

This arrangement provides an additional reduction of the dust and solids carryover from the shaft reactors to the fuel gas since no shaft reactor is operated to clean or desulfurize fuel gas in those intervals during which their burdens are being replenished.

In any of the above discussed embodiments, the refractory materials comprising burden 26 may be alumina spheres having a diameter greater than one half inch. Materials of this size prevent plugging of the burden 26 and can be easily separated from dust and other contaminants adhering thereto by a conventional screening process in station 110 while the refractory materials are hot. Recycling the refractory pellets while hot conserves energy, as

previously discussed.

Summary of the achievement of the objects of the invention

From the foregoing, it is readily apparent that I have provided an economical and energy efficient method for removing solids and sulfur from a hot fuel gas, which utilizes shaft reactors.

Since those skilled in the art can readily determine other possible alternative embodiments, this invention is to be limited only by the scope of the following claims.

CLAIMS

1. A method for cleaning a fuel gas in which the fuel gas passes through a first burden in a first shaft reactor to provide gross solid removal, through a second burden in a second shaft reactor to provide sulfur removal and fine solid removal, comprising the steps of:
 - (a) introducing fuel gas into a first shaft furnace having a packed bed burden therein, said burden including pellets of refractory material;
 - (b) passing said fuel gas through said burden to cause solids in said fuel gas to adhere to the surface of said pellets of refractory material;
 - (c) introducing said fuel gas into a second shaft reactor having a second burden therein, including a sulfur removing agent;
 - (d) passing said fuel gas through said sulfur removing agent to cause sulfur components of said gas to react with said sulfur removing agent to remove sulfur from said fuel gas; and
 - (e) removing said dedusted, desulfurized fuel gas from said second reactor.
2. A method for cleaning a fuel gas in accordance with claim 1 further comprising repeatedly removing portions of said hot refractory pellets comprising said burden, removing solids adhering to the surface thereof and recycling said portions of said hot refractory pellets to replenish the burden of said first reactor to reduce the loss of energy associated with replenishing said first burden.
3. A method for cleaning a fuel gas in accordance with claim 1 or 2, further comprising the additional step of introducing said gas into a cyclone cleaner to provide an additional stage of solid removal.
4. A method for cleaning a fuel gas in accordance with claim 1, 2 or 3, further comprising mixing limestone and iron oxide in predetermined ratios to form at least part of said second burden.
5. A method for cleaning a fuel gas in accordance with claims 3 and 4, further comprising introducing limestone into said cyclone to remove additional sulfur from said fuel gas.
6. A method of cleaning a fuel gas in accordance with claim 5, further comprising discharging and separating portions of said second burden to recover metallized iron and waste.
7. A method according to any preceding claim, wherein said sulfur removing agent has an average particle diameter of at least 0.5 centimeters.
8. A method according to any preceding claim, wherein said sulfur removing agent has an average

particle diameter of less than 3.0 centimeters.

9. A method for cleaning a fuel gas in accordance with any preceding claim, wherein said first burden includes limestone, dolomite, or a mixture thereof.

5 10. Apparatus for cleaning and desulfurizing a fuel gas stream, comprising in combination:

(a) apparatus for removing solids from said fuel gas stream comprising a first shaft reactor into which said fuel gas stream is introduced in

10 counterflow relation to a burden comprising materials selected to promote adhesion between solids contained in said fuel gas stream and said burden materials;

(b) means for recycling a portion of the materials comprising said burden whereby solids adhering to the recycled burden materials are removed and energy losses resulting from cooling of the recycled burden materials are reduced;

(c) means for directing said fuel gas stream through a second shaft reactor to react sulfur contained in said fuel gas stream with burden materials to produce solids containing sulfur, thereby removing sulfur from said fuel gas stream.

25 11. Apparatus for cleaning and desulfurizing a fuel gas stream in accordance with claim 10, wherein said burden includes refractory pellets of a predetermined size.

12. Apparatus for cleaning and desulfurizing a fuel gas stream in accordance with claim 11, wherein

30 said refractory pellets are alumina spheres.

13. Apparatus for cleaning and desulfurizing a fuel gas stream in accordance with claim 11 or 12, wherein said refractory pellets have a diameter of at least one centimeter.

35 14. The methods for cleaning a fuel gas substantially as hereinbefore described.

15. Apparatus for cleaning a fuel gas, substantially as hereinbefore described with reference to the accompanying drawings.

40